

# Temporal changes of $^7\text{Be}$ and $\text{PM}_{10}$ concentrations in surface air at (36.7°N 4.5 W)

C. Dueñas<sup>\*1</sup>, M.C. Fernández, E. Gordo, S. Cañete and M. Pérez<sup>2</sup>

<sup>1</sup>Department of Applied Physics I Faculty of Science.

<sup>2</sup> Department of Radiology and Health Physics, Ophthalmology and OTL. Faculty of Medicine.

Faculty of Science, University of Malaga, 29071 Malaga, Spain.

**Abstract.** Levels of particulate matter fraction  $\text{PM}_{10}$  were monitored between 2005 and 2009 in Málaga (Spain). The station “Carranque” (4° 28’ 4” W; 36° 43’ 40” N), belongs to the Atmospheric Pollution Monitoring network managed by the Environmental Health Service of the Andalusian Government.  $\text{PM}_{10}$  concentrations were measured at “Carranque” station by the beta attenuation method. The  $^7\text{Be}$  concentrations in air was continuously monitored, using an air sampler (Radeco, mod. AVS-28A) at a flow rate of 40 l/min and a high-resolution gamma-ray spectrometer, at the University of Málaga, (4° 28’ 4” W; 36° 43’ 40” N). Long-term measurements of cosmogenic radionuclides such as  $^7\text{Be}$  provide important data in studying of global atmospheric processes and comparing environmental impact of radioactivity from man-made sources to natural ones. The period of measurements was performed from 2005 to 2009. The variation of the data with time was studied by time series analyses and seasonal patterns were identified. The concentrations of  $^7\text{Be}$  exhibited maximum specific activities in spring and summer and one minimum in winter. The maximum concentrations for  $\text{PM}_{10}$  were observed, normally, in summer. Plots of the frequency distribution show highly skewed (flat on the right) histogram for  $\text{PM}_{10}$  and a symmetric for  $^7\text{Be}$ . The concentration data of  $^7\text{Be}$  and  $\text{PM}_{10}$  with meteorological variables were correlated to understand the monthly variation of these radionuclides in air. A complex relationship was observed between  $\text{PM}_{10}$  and  $^7\text{Be}$  concentrations in the measured aerosol filters collected at this site. Due to this fact, the analysed atmospheric events had to be grouped in: a) high  $^7\text{Be}$  and high  $\text{PM}_{10}$  matter events. b) Low  $^7\text{Be}$  and high  $\text{PM}_{10}$  matter events and c) high  $^7\text{Be}$  and low  $\text{PM}_{10}$  matter events. This study has shown that although  $^7\text{Be}$  and  $\text{PM}_{10}$  are associated with different source in Malaga, they may reach high concentration simultaneously. The reason for this is the concurrent concurrence of subsidence processes over North Africa (resulting in the downward transport of  $^7\text{Be}$  from the mid-troposphere) and the suspension of mineral dust over desert region with a subsequent transport to Malaga.

## INTRODUCTION

$^7\text{Be}$  (half life=55.3 days) is produced in the stratosphere and upper troposphere by the spallation of oxygen and nitrogen nuclides and is subsequently absorbed on to aerosols. Approximately 70% of  $^7\text{Be}$  is produced in the stratosphere, with the remaining 30% produced in the troposphere. Most of the  $^7\text{Be}$  that is produced in the stratosphere does not reach the troposphere except during spring, when the seasonal thinning of the tropopause takes place at midlatitudes, resulting in air exchange between the stratosphere and the troposphere. Since  $^7\text{Be}$  is of cosmogenic origin, its flux to the earth’s surface has a latitudinal dependence.

Its concentration in the air increases with increasing altitude from the surface of the earth, and its atmospheric flux to the earth’s surface should be independent of local land masses at any particular latitude (Baskaran et al., 1993).  $^7\text{Be}$  rapidly associates with submicron-sized aerosol particles. Gravitational settling and precipitation processes largely accomplish transfer to the surface of the earth.  $^7\text{Be}$  has become recognized as a potentially powerful tool when studying the description of environmental processes such as precipitation, wash-out (precipitation scavenging), atmospheric particle deposition and deposition patterns

of airborne contaminants (Papastefanou and Ioannidou, 1991). It is for this reason that  $^7\text{Be}$  has been, frequently, used as a tracer of stratospheric intrusions of gases and aerosols into the troposphere. However, there are few available studies on the impact of the suspension and transport of continental aerosols on the concentration of this radiotracer in the troposphere. But, the accurate knowledge of the local atmospheric deposition of  $^7\text{Be}$ , including the possible temporal variations produced by African dust events, could provide additional and at the same time, very useful, time markers in regional environmental historical archives.

The  $^7\text{Be}$  and PM<sub>10</sub> concentration measurements were carried out for the following reasons:

- 1) To know the temporal variation in the concentrations of  $^7\text{Be}$  and PM<sub>10</sub> in Malaga in order to determine whether they are affected in a similar way.
- 2) To determine the factor influencing the PM<sub>10</sub> concentration in the  $^7\text{Be}$  concentration in surface level air over the city of Malaga.

## **MATERIAL AND METHODS**

The site where the  $^7\text{Be}$  measurements were carried out in Malaga (4° 28' 4'' W; 36° 43' 40'' N) is in the North-West of the city, 5km away from the coastline. The sampling point (SP) was located on the roof of the Faculty of Sciences Figure 1 shows a map of Spain schematically and the sampling point location. Malaga is the major coastal city of Andalusia region, South Spain. This Spanish city on the Mediterranean is distinguished by its mild temperate and warm climate with low rainfall (550 mm $\text{yr}^{-1}$ ) and around 320 days of sun a year. The coast is backed by a series of mountains that have to be crossed to reach the inland valleys.

As Malaga is located on the coast, its ambient air is influenced by both continental and maritime air masses. Due to the influence of the local orography, SE and NW winds prevail and these winds can be observed in the sea-land and land-sea breezes, respectively.

Airborne dust samples were collected weekly in cellulose nitrate filter, 47 mm diameter (collection efficiency 99.99% for 0.8  $\mu\text{m}$  pore size) with an air sampler (RADECO, model AVS-28A) at a flow rate of 40 l  $\text{min}^{-1}$ , covering a total period from October 2007 to October 2009. Sample air volumes were determined by an in-line dry gas meter and about 300 m<sup>3</sup> atmospheric air per week have been drawn through the filter for the routine sampling that has been performed. The location and positioning of this aerosol pump a 12 m was chosen to minimise the contamination of the filters by re-suspension of local soil particles.

Measurements by gamma-spectrometry were performed to determine the  $^7\text{Be}$  activities of the samples using an intrinsic REGe. The measurements could perform on single filter determination. The  $^7\text{Be}$  concentration was calculated using the 477.6 keV gamma-ray line, the counting time was 345,600 s. The reported uncertainty is propagated errors arising from the one sigma counting uncertainty due to detector calibration and background correction. The concentrations were corrected for decay to the mid-collection period.

PM<sub>10</sub> concentrations were measured every ten minutes at "Carranque" station (CS) by the beta attenuation method. "Carranque" station (4° 26' 3" W; 36° 40' 29" N), belongs to the Atmospheric Pollution Monitoring network managed by the Environmental Health Service of the Andalusian Government.



Figure 1. The location of sampling point (SP) and Carranque station (CS).

## RESULTS AND DISCUSSION

The  $^7\text{Be}$  dates used in the analysis were monthly values of concentration in surface air and were carried out from October 2005 to October 2009.

The results from individual measurements of  $^7\text{Be}$  and  $\text{PM}_{10}$  concentration were analyzed to derive the statistical estimate characterizing the distribution. Table 1 provides arithmetic mean (AM) and related statistical information such as geometric mean (GM), standard deviation (SD), dispersion factor of geometric mean (DF), maximum and minimum value. These values are given in  $\text{Bq m}^{-3}$  and  $\mu\text{g m}^{-3}$  respectively.

The  $^7\text{Be}$  dates in the period of measure were analyzed to derive the statistical estimates characterizing the distribution. Studies of the frequency distribution show lognormal distribution with 0.01 significant levels.

	AM	GM	SD	DF	Maximum	Minimum
$^7\text{Be}$ ( $\text{Bq m}^{-3}$ )	$4.3 \cdot 10^{-3}$	$3.7 \cdot 10^{-3}$	$1.7 \cdot 10^{-3}$	1.82	$8.1 \cdot 10^{-3}$	$7.2 \cdot 10^{-4}$
$\text{PM}_{10}$ ( $\mu\text{g m}^{-3}$ )	27.5	22.7	18.1	1.88	84	5

Table 1. Statistical parameters of the different measurements

The Figure 2 shows the monthly mean  $\text{PM}_{10}$ , and  $^7\text{Be}$  concentrations. As it can be observed in this figure, the relationship between  $^7\text{Be}$  and  $\text{PM}_{10}$  is rather complex and variable. See how high  $^7\text{Be}$  events occurred during either high or low  $\text{PM}_{10}$  episodes. Moreover, the figure shows the seasonal variations, with two peaks in summer and fall in winter in the period Oct-05 to Apr-08, this is a typical seasonal profile for Malaga (Dueñas et al 1999; Dueñas et al. 2004) with average values low  $\text{PM}_{10}$  of  $19 \mu\text{g m}^{-3}$ . On the other hand, we can observe like the figure shows low  $^7\text{Be}$  and high  $\text{PM}_{10}$  events in the spring -summer period from May-08 to Oct-09.

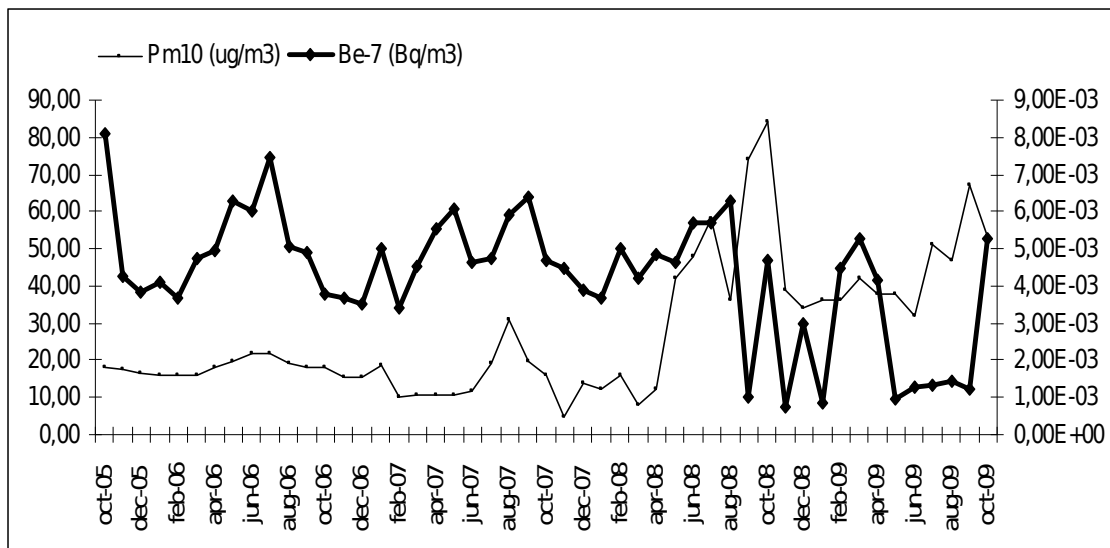


Figure 2. Variations in  $^7\text{Be}$ ,  $\text{PM}_{10}$  concentrations in air

In other hand, in order to simplify the analysis, the events have been grouped into two period:

- (1) Low  $\text{PM}_{10}$  in the period Oct-05 to Apr-08.
- (2) High  $\text{PM}_{10}$  in the period May-08 to Oct-09.

In events of high  $^7\text{Be}$  and low  $\text{PM}_{10}$ , the values for  $^7\text{Be}$  oscillated between  $3.41 \cdot 10^{-3}$  and  $8.12 \cdot 10^{-3} \text{ Bq m}^{-3}$  with a mean value of  $4.88 \cdot 10^{-3} \text{ Bq m}^{-3}$ . The values for  $\text{PM}_{10}$  oscillated between  $30.71$  and  $5 \mu\text{g m}^{-3}$  with a mean value of  $15.92 \mu\text{g m}^{-3}$ .

As meteorology plays an important role in the dispersion and transport of pollutants, the study of correlation between  $^7\text{Be}$  and meteorological parameters reveals a pronounced correlation with air temperature (Dueñas et al., 2005). High temperatures are often associated to upward convection currents in the atmosphere. Several events of high  $^7\text{Be}$  concentrations are mainly caused by downward transport of  $^7\text{Be}$  from the midtroposphere at mid-latitudes have been identified during the study period. Some examples are highlighted October 2005, July 2006 and May 2007. During those events, relatively high  $^7\text{Be}$  concentration and low  $\text{PM}_{10}$  concentration are produced. During October 2005, July 2006 and May 2007 a cut-off low event developed over central Europe (Bonasoni et al., 2000). The meteorological situation was characterised by an Atlantic anticyclone system with a low-pressure area over central European North Atlantic. (Prospero et al., 1995) also found high  $^7\text{Be}$  concentrations associated with low aerosol mass concentrations. These values were attributed to downward transport from mid-to-upper troposphere over the North Atlantic (Figure 3).

The meteorological situation was characterised by a south Europe anticyclone located in the South Atlantic Ocean that favoured the development of a blocking system. Under these conditions, a high pressure area can be developed over the North Atlantic (Hernandez et al., 2005).

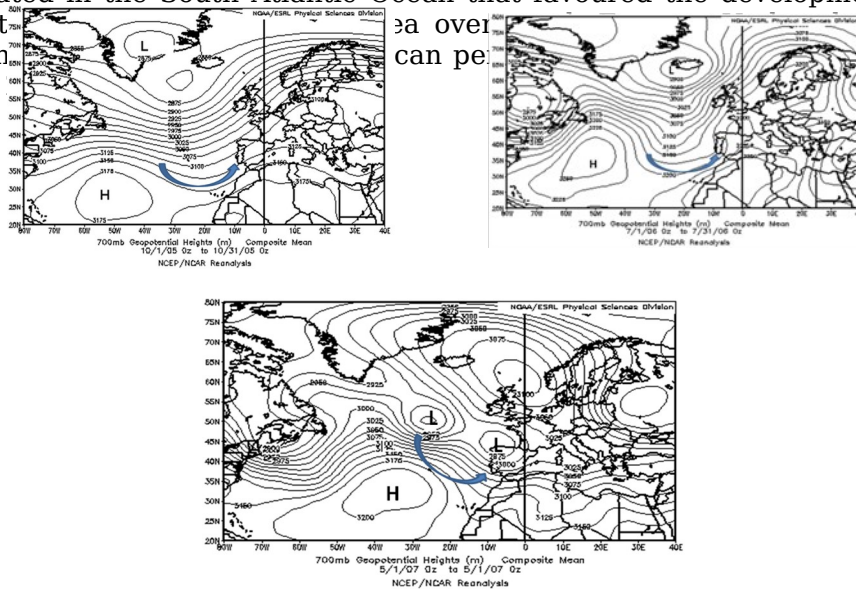
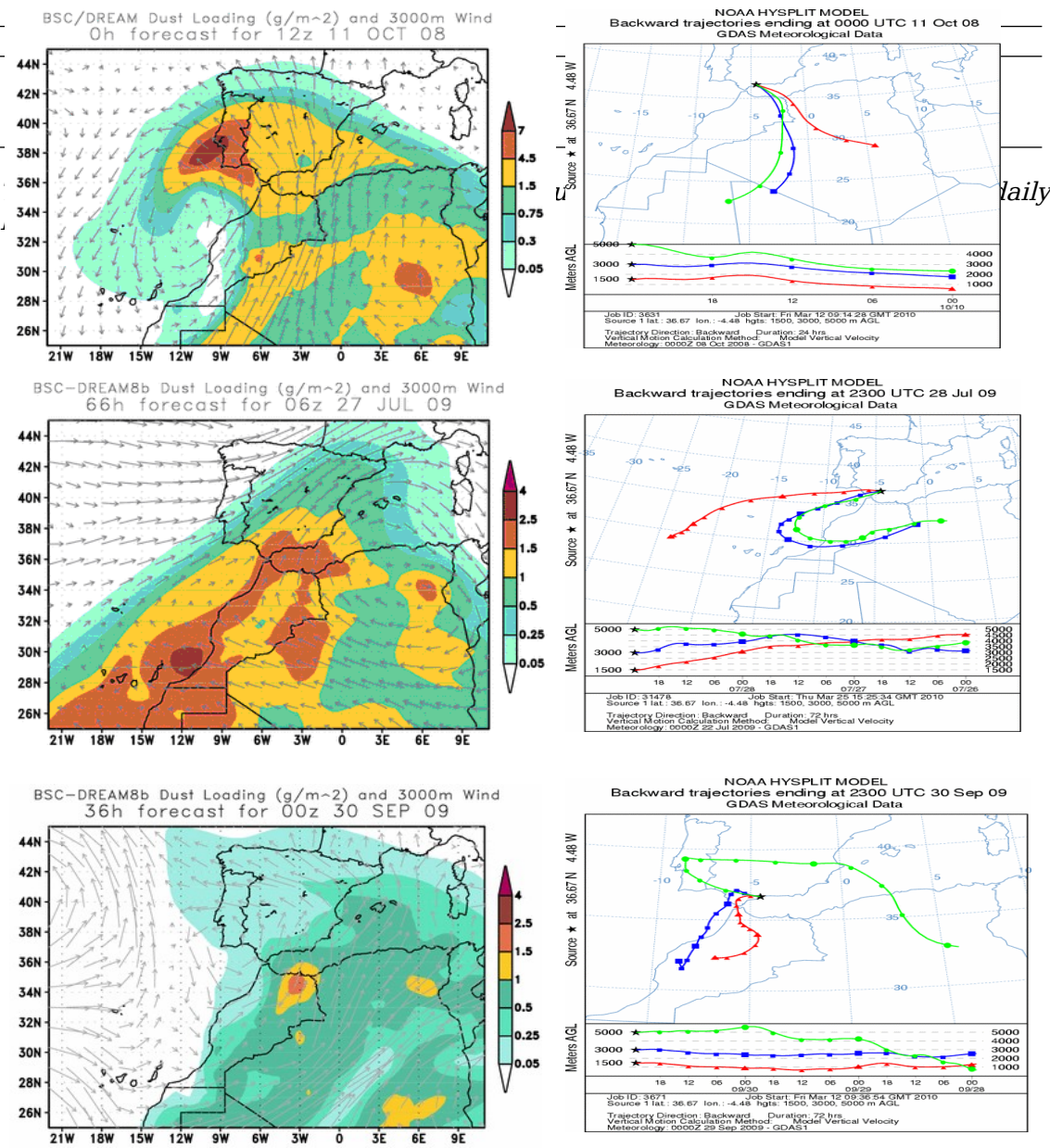


Figure 3. Geo-potential surface altitude during the high <sup>7</sup>Be and low PM10

In events of low <sup>7</sup>Be and high PM<sub>10</sub>, in the period May-08 to Oct-09 the values for <sup>7</sup>Be activity oscillated between  $7.21 \cdot 10^{-4}$  to  $6.26 \cdot 10^{-3}$  Bq/m<sup>3</sup> with a mean value of  $3.22 \cdot 10^{-3}$  Bq/m<sup>3</sup>. A range of 52 µg/ m<sup>3</sup> and mean value of 47.50 µg/ m<sup>3</sup> at ground level air were measured for PM10 concentration. According to the EU framework directive 1999/30/EC the limit value for daily PM10 average is 50 µg/m<sup>3</sup> and must not be exceeded on more than 35 days of the year (valid for years 2005-2009). However, this Directive accepts that countries will sometimes be subject to PM<sub>10</sub> pollution events ascribable to natural events such as the atmospheric re-suspension or transport of natural particles from dry regions.

In the table 1, there are highlighted mean monthly values of PM10 concentrations in October 2008, July 2009 and September 2009. The table 2 provides arithmetic means, maximum values and % maximum daily limit values exceeded.



*Figure 4. Map of atmospheric dust loads and back-trajectories*

PM<sub>10</sub> levels are, also, very useful markers to characterise Saharan dust intrusions over the Malaga city. This paper study the origin of the air masses reaching the station “Carranque” when there is outbreak. In these episodes, the low <sup>7</sup>Be and high PM<sub>10</sub> concentrations were accounted by air masses originated and moving at low altitudes over North Africa (according to the performed back-trajectory analysis). Daily PM<sub>10</sub> levels recorded at regional background stations during this episode were high, causing PM<sub>10</sub> excess in Oct-08, Jul-09 and Sep-09. During these episodes Saharan intrusions over Iberian Peninsula were mostly induced by the simultaneous occurrence of a western/southwestern depression and eastern anticyclone (S. Rodriguez et al 2001).

Figure 4 shows the back-trajectories and map of atmospheric dust loads during a typical example of this type of events. Low <sup>7</sup>Be and high mineral dust concentrations events recorded during the summer in connection with Saharan dust inputs events agree with the observations performed by Prospero et al. (1995).

## CONCLUSIONS

As stated above (results and discussion), the time resolution of the radiometric data was 1 month. This is a relatively long period during which atmospheric transport patterns prompting different relationship between <sup>7</sup>Be concentrations and PM<sub>10</sub> may occur (even opposites ones). For that reason, the discussion is based on a set of very clear examples with dominant transport patterns during each sampling month.

A complex relationship was observed between PM<sub>10</sub> and <sup>7</sup>Be concentrations in the measured aerosol filters collected at this site. This study has shown that although <sup>7</sup>Be and PM<sub>10</sub> are associated with different source in Malaga, they may reach high concentration simultaneously. The reason for this is the concurrent concurrence of subsidence processes over North Africa (resulting in the downward transport of <sup>7</sup>Be from the mid-troposphere) and the suspension of mineral dust over desert region with a subsequent transport to Malaga.

#### **4. References.**

- Bonasoni P., Evangelisti F., Bonafe U., Ravegnani F., Calzolari F., Stohl A., Tositti L., Tubertini O. and Colombo T., 2000. Stratospheric ozone intrusion episodes recorded at Mt. Cimone during the VOTALP project: case studies. *Atmospheric Environment* 34, 1355-1365.
- Dueñas C., Fernandez M.C., Liger E. and Carretero J., 1999, Gross alpha, gross beta activities and  $^7\text{Be}$  concentrations in surface air: analysis of their variations and prediction model. *Atmospheric Environment* 33, 3705-3715.
- Dueñas C., Fernandez M.C., Liger E., Carretero J. and Cañete S., 2004, Long-term variation of the concentrations of long-lived Rn descendants and cosmogenic  $^7\text{Be}$  and determination of the MRT of aerosols. *Atmospheric Environment* 38, 1291-1301
- Rodriguez S., Querol X., Alastuery A., Kallos G., Kakaliago O., 2001. Sahara dust contributions to PM<sub>10</sub> and TSP levels in Southern and Eastern Spain. *Atmospheric Environment* 33, 2433-2447.
- Hernandez F., Rodriguez S., Karlssona L., Alonso-Perez S., Lopez-Pereza M., Hernandez-Armas J. and Cuevas E., 2008, Origin of observed high  $^7\text{Be}$  and mineral dust concentrations in ambient air on the Island of Tenerife. *Atmospheric Environment* 42, 4247-4256
- Prospero J.M., Schmitt R., Cuevas E., Savoie D.L., Graustein W.C., Turekian K.K., Volz-Thomas A., Díaz A., Oltmans S.J. and Levy H., 1995. Temporal variability of summer-time ozone and aerosols in the free troposphere over the eastern North Atlantic. *Geophysical Research Letters* 22 (21), 2925-2928